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### COSMOGENIC NUCLEI

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#### 1. INTRODUCTION

Cosmogenic nuclei are, by definition, nuclides formed by nuclear interactions of galactic and solar cosmic rays with extraterrestrial (meteorites, moon, interplanetary dust, etc.) or terrestrial (atmosphere, lithosphere, etc.) matter. The nuclides produced in these reactions range from short lived radioactive species to stable isotopes. In this paper we will, for two reasons, concentrate on the long lived ( $\sim 10^2$ - $10^7$  years) radioactive cosmogenic isotopes. First, it is these isotopes which remain in various geological reservoirs today, as a link with cosmic ray activity in the past. Unlike stable cosmogenic nuclei (with some important exceptions) these long lived isotopes can readily be distinguished from "ordinary" terrestrial matter, and thus are unambiguous evidence of cosmogenic production. The second reason is that the study of these long lived species has been revolutionized (and the word is not too strong) by the development in the past few years of a technique known as accelerator mass spectrometry (AMS). It is in fact the participation of our group at Orsay in the development of this technique, together with our previous interest in cosmic rays, which has led to our involvement in the study of cosmogenic nuclei.

We will not here go into any details about the technique of AMS, but rather refer the interested reader to the proceedings of the last symposium on this subject (1). Basically AMS is mass spectrometry at an energy where nuclear as well as atomic forces can be exploited in the

separation and detection steps. The result is a technique which can sensitively, and uniquely, identify a very small quantity ( $\sim 10^6$  atoms) of a given isotope in a much larger matrix of other nuclei. It is this property which makes it particularly valuable for measuring the small concentration of cosmogenic nuclei often available in geological samples.

AMS was originally developed at accelerators built for, and largely devoted to, nuclear physics. Much current work continues to be done at such accelerators. However, the potential of the technique has also led to the design and installation of five so-called "dedicated" accelerators, at Oxford University, University of Arizona, University of Toronto, Nagoya (Japan) and Gif-sur-Yvette (France). These relatively small ( $\sim 2$  million volt terminal voltage) tandem accelerators are used full time for AMS. Although originally conceived for  $^{14}\text{C}$  measurements, we have shown that these Tandetron accelerators are also capable of measuring  $^{10}\text{Be}$  and  $^{26}\text{Al}$  with sensitivity and background levels comparable to the higher energy machines (2) (3). Except where noted, the results mentioned in the present paper have been obtained using the Tandetron accelerator at Gif-sur-Yvette.

The range of applications of cosmogenic isotopes is much too large to cover in any detail here. Once again, a simple perusal of ref (1) will give the interested reader some idea of the breadth of these applications. Thus, after briefly categorizing the types of these applications, I choose to describe several recent studies undertaken by our own group, which I feel might interest the attendees of this meeting. This paper is thus in no way intended to be a comprehensive or general review. The choice of subject matter is one of convenience and topicality and does not reflect any value judgement on similar or different studies being carried out by other groups.

## 2. GENERAL APPLICATIONS

We like to classify the applications of cosmogenic nuclei into three broad categories :

a) Dating : this is perhaps the first application that comes to mind when thinking of radioactive isotopes. With the exception of the already well developed procedure of  $^{14}\text{C}$  dating, however, it is the application which will probably require the most extensive and detailed preliminary studies before it can be fully exploited. The reason is that, unlike  $^{14}\text{C}$ , most of the other cosmogenic isotopes are not homogenized with their stable isotopes in the atmosphere. Their application in dating thus necessitates, at the very least, a detailed and comprehensive knowledge of the geochemistry of the isotope in question. In some cases application may require such limiting conditions as to be impractical.

b) Tracers : as mentioned above, cosmogenic nuclei are unambiguous witnesses of the interaction of cosmic rays with matter. As such they can be thought of as "tracers" of these interactions, and be used in two general ways :

- First, they can give information on the duration, place and conditions of the irradiation

- Second, once formed, they can give information on the movement of the medium (air, water, soil, ice, etc) in which they are transported.

c) Production variations, and their implications : there are basically three parameters which control cosmogenic production rates

- i) Primary galactic cosmic ray intensity

- ii) Solar activity (through modulation and production of solar flare particles)

- iii) Geomagnetic field intensity (for terrestrial production)

The study of cosmogenic isotopes as a function of stratigraphic position in various geological reservoirs (marine and lacustrine sediments, polar ice) can thus potentially give us information on the variation of the above three parameters in the past.

### 3. SOME RECENT EXAMPLES

a) Primary cosmic rays : the application which probably most directly interests attendees at this meeting is the possibility of obtaining information on the variation of primary galactic cosmic ray flux in the past. Many theories of cosmic ray formation and acceleration allow, or even predict, a variable flux at the earth on a geological time scale. For example, at the Paris Cosmic Ray Conference, Axford discussed the type of variability that might be expected from the acceleration of cosmic rays by shock waves associated with supernova remnants (4). Streitmatter et al. (5) have recently described the type of variations that could result from the acceleration of cosmic rays in a "superbubble" in which the solar system is presently imbedded. At the present meeting Wolfendale et al. (OG 3.1-11) have given their predictions on the expected variability of intensity of cosmic rays associated with supernova remnants. In each of these cases, the variability is expected to be quite significant over periods of the order of  $\sim 10^4$ - $10^7$  years. This is just the period which is amenable to study using the nuclide  $^{10}\text{Be}$  (half-life 1.5 My) which, conveniently, is also the second most abundant cosmogenic nuclide, after  $^{14}\text{C}$ , produced in the earth's atmosphere.

Already studies on meteorites, and a few limited measurements in marine sediments, using classical counting techniques, have permitted tentative limits on cosmic ray variability (see, for example, Foreman and Schaeffer (6), and Reedy et al. (7) and references therein). However, the technique of AMS offers the promise of much more extensive and detailed limitations in the near future. At Orsay we are working on such an investigation by measuring  $^{10}\text{Be}$  concentrations in several marine sediment cores. We have previously reported a few results from marine core RC12-65, which suggested a possible increase in production  $\sim 10$  My ago (8). We have since measured a substantially larger number of samples from this core. In addition, the chronology of the magnetic stratigraphy used to date the core has recently been subject to revision (9). This has had the effect of significantly modifying the age of the samples in

the ~ 5-9 My time range, and thus the earlier conclusions. We show in Fig. 1 some new results, expressed as  $^{10}\text{Be}/^9\text{Be}$  ratio. In fact, one of the most difficult aspects in using  $^{10}\text{Be}$  to deduce production variations, is the problem of how to normalize the results. Although we do not believe that  $^{10}\text{Be}$  and  $^9\text{Be}$  are completely homogenized in the ocean, we have evidence that, in some cases at least,  $^9\text{Be}$  can partially compensate for varying sediment composition (in particular, the biogenic component). Thus  $^9\text{Be}$  can serve as a useful, although imperfect, normalizing species.

Each sample in Fig. 1 represents ~ 1 cm depth in the core. However, bioturbation (mixing by organisms at the sediment surface) typically mixes deep sea sediments over a depth of ~ 10 cm. Using the sedimentation rates determined in the core by magnetic stratigraphy, a 10 cm depth interval corresponds to ~ 80,000 years in the upper part of the core, and ~ 20,000 years toward the bottom. This then determines the time resolution of the  $^{10}\text{Be}$  measurements.

For the moment we have not included in Fig. 1 the earlier data of Ref (8). The reason is that the  $^9\text{Be}$  measurements in that work were made under slightly different conditions, and we have not yet confirmed the reproducibility of the two techniques. The errors in Fig. 1 have been calculated using, in addition to the uncertainty in the  $^{10}\text{Be}$  measurements, a 7 % relative uncertainty for the  $^9\text{Be}$ . This is the average  $^9\text{Be}$  variability observed in a series of duplicate measurements in another core (10). However it is possible that additional measurements in the present core may lead to revisions outside this range. Ironically enough, we presently have more confidence in the precision of our  $^{10}\text{Be}$  measurements than those of the  $^9\text{Be}$ .

For a perfectly constant cosmogenic production rate, and uniform sedimentation conditions, the  $^{10}\text{Be}/^9\text{Be}$  ratio in Fig. 1 should decrease with age in the sediment with the 1.5 My half-life of  $^{10}\text{Be}$ . Although there are some minor deviations, the most notable aspect of the data is the degree to which they follow such a trend. In addition to possible production variations, the deviations which do exist could be due to experimental error, rapid changes in sedimentation conditions, or

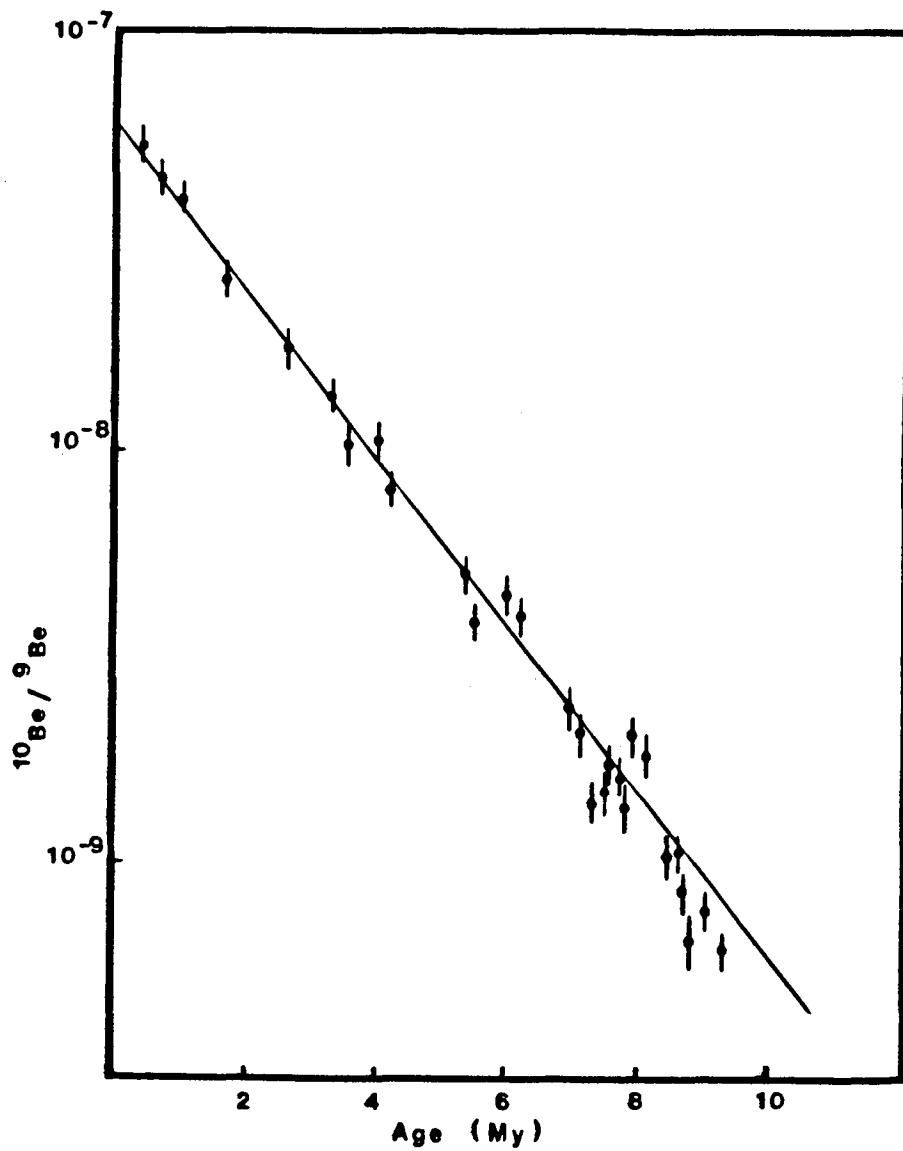


Fig 1 :  $^{10}\text{Be}/^9\text{Be}$  ratio as function of age of samples  
in marine sediment core RC12-65

residual uncertainties in the chronology of the core. In order to check for such possibilities it will be necessary to make similar measurements in other cores covering the same time period.

Even if production variations are established it will be necessary, before concluding that these are due to primary cosmic ray intensity changes, to consider two other potential sources of variation mentioned above, namely solar modulation and geomagnetic field variation. The maximum expected variation due to these causes is a factor of  $\sim 2$ , and they are expected to be significantly attenuated over time periods of  $\sim 10^4$ - $10^5$  years (ie the time resolution of the samples in Fig. 1). Thus the type of variations which would most strongly suggest a primary cosmic ray origin would be those of large ( $> \sim 2$ ) amplitude, or long ( $> \sim 1$  My) duration.

Within the uncertainties and time resolution of the data, the measurements shown in Fig. 1 provide no compelling evidence for changes in cosmic ray flux during the past  $\sim 9$  My (although there is a hint of a brief increase at  $\sim 8$  My). This conclusion is similar to that arrived at by Tanaka and Inoue (11) over a shorter time period (2.5 My) or Ku et al. (12) over approximately the same time period, but with poorer time resolution. However, I wish to emphasize that the results of Fig. 1 are still preliminary, and incomplete. In addition to making more detailed measurements in RC12-65, we are extending this study back to  $\sim 20$  My by making similar measurements in other cores. The purpose in presenting the results of Fig. 1 is thus not to give here any definitive limits to possible cosmic ray variations, but rather to illustrate the type of data that can be expected in the quite near future. Such data may then provide important constraints to theories of the origin and acceleration of cosmic rays. I would thus urge those working on such theories to try to calculate and report the time variations to be expected from their models.

b) Variation at a geomagnetic reversal : The intensity of the geomagnetic field varies on time scales of hundreds to tens of thousands of years (13) (at the present rate of change, the field would become zero in about 2000 years ). The most dramatic of these changes occurs during a geomagnetic reversal. During such an event it is believed that the dipole field intensity decreases to  $< 20\%$  of its "normal" value, for a period of the order of  $10^4$  years. During such a time, we would thus predict that the production rate of cosmogenic nuclides in the atmosphere should increase. In order to test this idea, and obtain additional information on the details and length of the intensity changes, we have measured a  $^{10}\text{Be}$  profile in a marine sediment core during the most recent of these reversals (Brunhes-Matuyama), which occurred 730,000 years ago. The results, shown in Fig. 2, do indeed show a significant increase in  $^{10}\text{Be}$  at the time of the reversal (10). This increase in production occurs over a period estimated as  $\sim 12,000\text{--}24,000$  years, and is considerably longer than the change in direction itself. Further studies along these lines should help those working on models of the reversal process itself, and on the way the magnetic signal is acquired in marine sediments.

c)  $^{10}\text{Be}$  in polar ice cores : Some of our first measurements of  $^{10}\text{Be}$  by AMS were made on samples from an Antarctic ice core. One of the principle motivations for that work was to look for variations caused by variable solar modulation. We did indeed find increased  $^{10}\text{Be}$  during the period 1645-1715 AD, known as the Maunder Minimum (14). Somewhat more surprising, we also found increased  $^{10}\text{Be}$  concentrations in ice deposited during the last ice age. More recent work by a Swiss collaboration, using ice cores from Greenland, has confirmed and extended these observations (15) (16). Nevertheless, the interpretation of the increase during the ice age has remained uncertain. We have recently had the opportunity to measure  $^{10}\text{Be}$  in a 2083 m core taken in Antarctica by a Russian group. This core goes back in time over the entire last climatic cycle ( $\sim 125,000$  years). We once again found increased  $^{10}\text{Be}$  concentration during the glacial stages, with concentrations similar to the



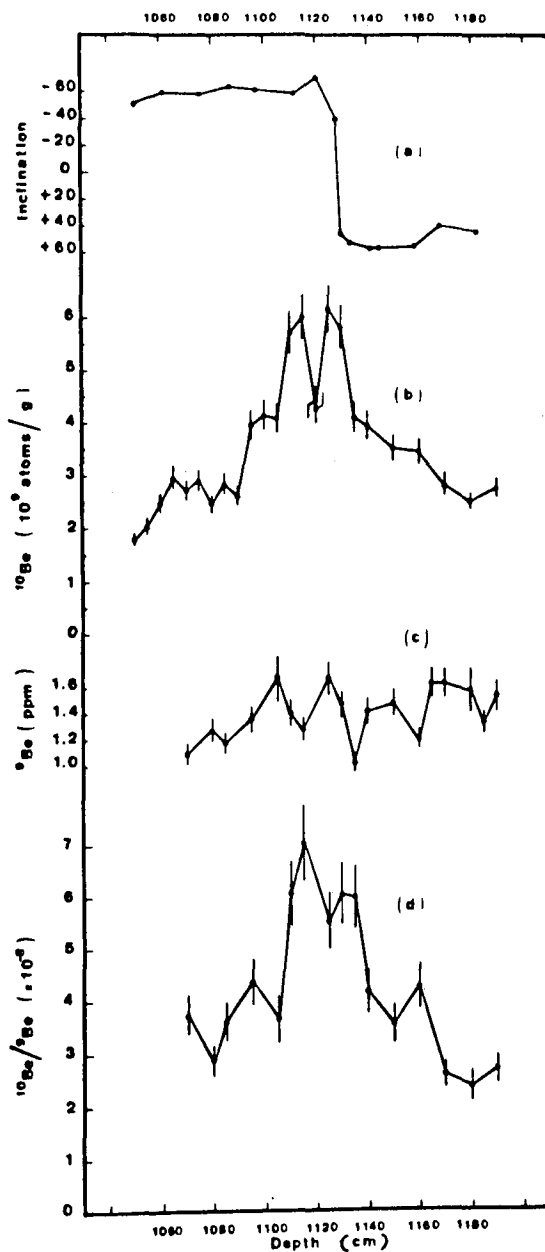


Fig 2 : (a) Magnetic inclination (b)  $^{10}\text{Be}$  concentration  
 (c)  $^9\text{Be}$  concentration (d)  $^{10}\text{Be}/^9\text{Be}$  ratio in marine  
 core V16-58 during geomagnetic reversal (from ref 10)

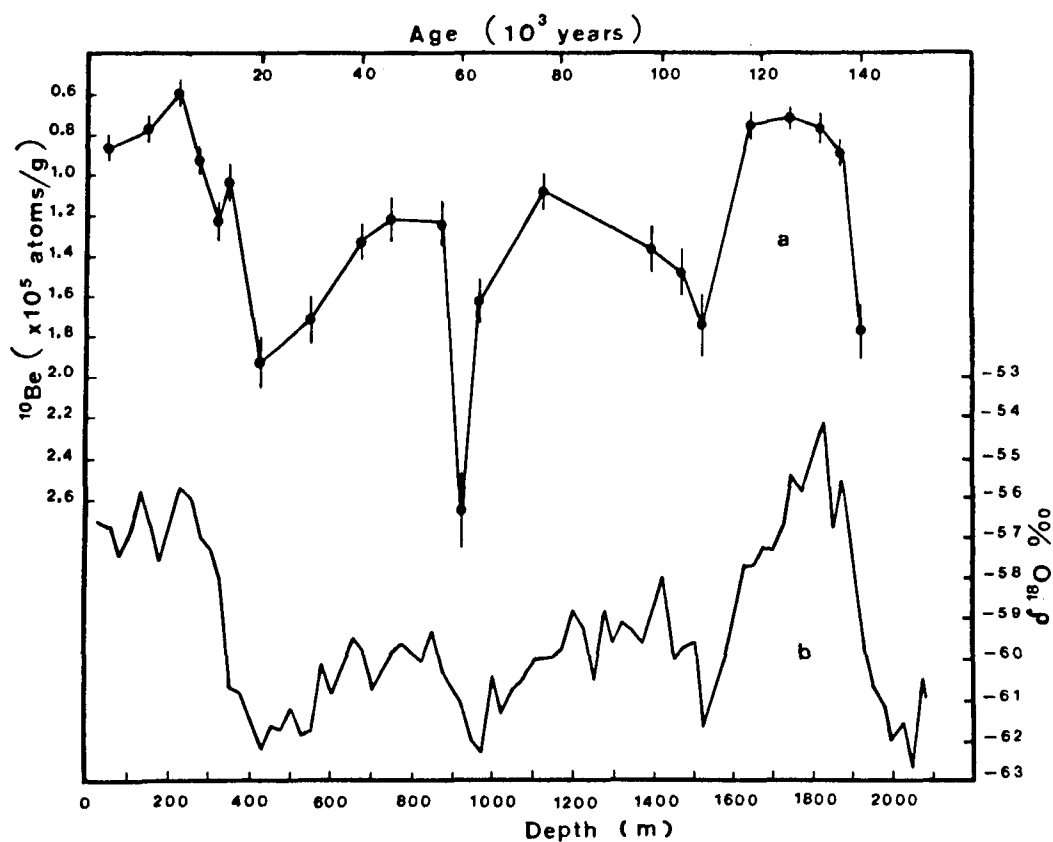


Fig 3 :  $^{10}\text{Be}$  concentration and  $\delta^{18}\text{O}$  in an ice core from Vostok station Antarctica (from ref 17) Note inverted scale of  $^{10}\text{Be}$  to facilitate comparison between curves

present during the last interglacial (17). Our present interpretation of this phenomena is that the changes are not due to  $^{10}\text{Be}$  production variations, but rather changes in the past precipitation rate in the Antarctic. Knowledge of such past precipitation rates is not only of interest for climatic studies, it is essential for determining the chronology of the ice cores themselves.

d)  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in cosmic spherules : Cosmic spherules are small (~ 50-500 micron diameter), magnetic objects originally found in slowly accumulating deep sea sediments (18). An origin as ablation products from extraterrestrial material during atmospheric entry was first suggested by their discoverers, more than 100 years ago (19). The exact nature of the parent bodies has, however, remained uncertain. Several years ago, Nishiizumi, on the basis of cosmogenic  $^{53}\text{Mn}$  data, suggested ablation from "normal" sized meteorites (20). An alternate possibility is that the parent objects could be the much more numerous small (~ 1 mg) objects that bombard the atmosphere (often observed as "shooting stars"), and that are believed to represent cometary debris.

The possibility of measuring cosmogenic  $^{10}\text{Be}$  and  $^{26}\text{Al}$  in the spherules suggested to us a way of distinguishing between these two possibilities. If the parent bodies were irradiated as small objects in interplanetary space, they should have a much larger  $^{26}\text{Al}/^{10}\text{Be}$  ratio than that found in larger meteorites. The reason is that, in addition to formation by galactic cosmic rays,  $^{26}\text{Al}$  can also be formed from the more numerous lower energy solar flare particles.  $^{10}\text{Be}$ , on the other hand, being formed most efficiently by higher energy reactions, will have only a modest contribution from solar flare particles. Since the lower energy solar flare particles have a relatively short range (few millimeters) in matter, their influence will be significant for only small parent bodies. Using the University of Pennsylvania tandem accelerator, we measured the  $^{26}\text{Al}/^{10}\text{Be}$  ratio in groups of (21), and individual (22) cosmic spherules. The ratios we found were in general much larger than found in meteorites, suggesting to us that these spherules are, in fact, in large part, cometary debris. Among our most recent measurements, made

with the Tandetron, we have found a  $^{10}\text{Be}$  concentration in one spherule which is much larger than the "saturation" value for irradiation in near earth interplanetary space (23). We have suggested that this spherule may have been irradiated in part outside the solar modulation region, where the galactic cosmic ray flux is believed to be much larger than observed in the interplanetary space explored so far.

We are looking forward to extending these studies to similar spherules recently discovered in "blue lakes" on Greenland ice (24). The collection procedure in this case does not depend on the spherules being magnetic, and they may include better preserved, and even new forms, of extraterrestrial matter.

#### 4. Conclusion

The technique of AMS opens up whole new areas of application for long lived cosmogenic nuclei. In addition to creating new links between cosmic ray physics and other domains, the possibility now exists for adding a virtually new dimension to cosmic ray studies themselves, namely that of detailed time variability over the past  $\sim 20$  My. Such information may well provide important implications for theories of cosmic ray origin and acceleration.

#### 5. Acknowledgements

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